

ARCO Chemical Company  
Legal Department  
Patent Section  
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Newtown Square, Pennsylvania 19073-2387  
Telephone (610) 359 2109  
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John C. Martin, Jr.  
Senior Patent Counsel

November 21, 1995

John Kollar  
President  
Redox Technologies Inc.  
6 Spencer Court  
Wyckoff, NJ 07481

Re: Redox's EG Technology

Dear Mr. Kollar:

I acknowledge receipt of your October 31 letter relating to Redox's EG Technology.

Following our October 31 meeting, we reviewed earlier technical suggestions and enclose for your information a copy of Technical Suggestion 81-73 signed October 16, 1981 titled "Improved Process for the Production of Ditertiary Butyl Peroxide." Preparation of DTBP is described by the reaction of either TBA or isobutylene using tertiary butyl hydroperoxide (TBHP) and an ion exchange acid resin catalyst. The first paragraph, second typed page of the Technical Suggestion states that "a large pore size resin would be preferable." As you know, large pore size resins are known as macroreticular resins.

Therefore, this Technical Suggestion meets the paragraph 6(b) exception to confidentiality of the April 23, 1987 agreement in that the information disclosed to ARCO Chemical Company was in its possession prior to receipt from Redox.

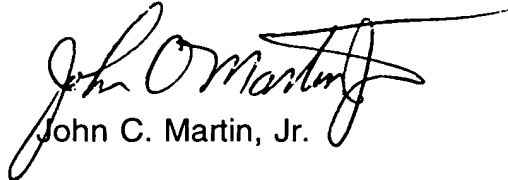
As I stated at our October 31 meeting, we are willing, at your request, to provide affidavits from all named inventors of our 5,371,298 patent stating that they did not have access to any of Redox's technology (i.e., their invention was independently developed from your information). See paragraph 6(d) of the April 23, 1987 agreement.

John Kollar  
November 21, 1995  
Page 2

Some of us believe that using macroreticular resins for this reaction is in the publicly available literature such as early Rohm & Haas Company product bulletins describing uses of macroreticular resins. To date, such literature has not been uncovered.

In any event, the information already provided to you by this letter and at our October 31 meeting clearly eliminates any possible claim by Redox against ARCO Chemical Company under the April 23, 1987 agreement.

Sincerely,



John C. Martin, Jr.

cc: W. J. Klingebiel

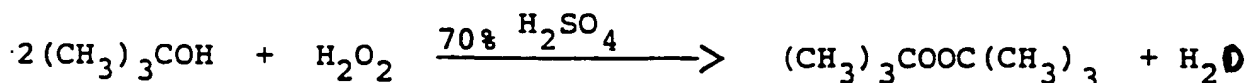
Enclosure

T. S. No. 81-13 Date Assigned 10/16/81

(1) Title of Invention Improved Process for the Production of Ditertiary  
\*Butyl Peroxide

(2) - (3) - (4) - (5)

Ditertiary butyl peroxide (DTBP) is a specialty product used primarily as an organic peroxide catalyst or catalyst intermediate. It is commercially produced by the reaction of pure t-butanol (TBA) or isobutylene with hydrogen peroxide in the presence of concentrated sulfuric acid:



The process requires refrigeration to control the reaction isotherm ( $\Delta H = 98,000$  Btu/lb mole) at or below  $15^\circ\text{C}$  thus reducing side reactions and TBA dehydration. The product mixture is quenched with excess water to promote phase separation. The organic phase, consisting of mostly DTBP, is washed with dilute alkali solution and the aqueous acid phase is usually neutralized and discarded.

When excess hydrogen peroxide is used in the above reaction, a mixture tert-butyl hydroperoxide and DTBP is obtained.

An alternative process, that has been experimentally demonstrated and recently evaluated (1), substitutes tert-butyl hydroperoxide for the hydrogen peroxide:

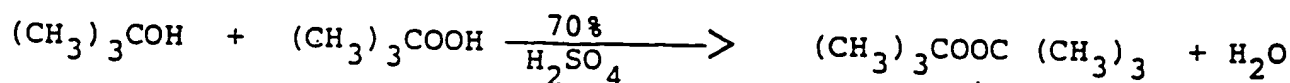
Project No. _____		(6) Test data and original references may be found on the following notebook pages <u>187434, 187435</u>	
Inventor (s) Signature E. A. Hazbun <i>E. A. Hazbun</i>		Date signed 10/13/81	Date of Conception
Signature of Witness V. M. Chong		Date signed 10/15/81	Date first disclosed to me
Signature of Witness J. A. Cahill		Date signed 10/15/81	Date first disclosed to me



T. S. No. 801Date Assigned 10/16/81

(1) Title of Invention Improved Process for the Production of Ditertiary Butyl Peroxide

(2) - (3) - (4) - (5)



This also requires refrigeration and acid neutralization. The estimated transfer price via this process at the 3 MM lbs/yr scale is \$1.16/lb for pure DTBP.

Recent preliminary testing of DTBP as a cetane improver for diesel fuels (2) have shown the following encouraging results:

V %	Cetane #
0	44.8
3	56.8
5	67.8

Such a performance may create a large volume application for DTBP if it can be manufactured at low cost.

To allow continuous production at low cost, I propose that DTBP be produced by the reaction of TBHP and TBA over an ion exchange acid resin catalyst at a temperature sufficiently elevated to permit cooling with cooling tower water obviating the need for refrigeration.

Project No. \_\_\_\_\_

(6) Test data and original references may be found on the following notebook pages 187435 187436

Inventor(s) Signature

E. A. Hazbun

Date signed

10/13/81

Date of Conception

Date signed

Date of Conception

Signature of Witness

V. M. Chong

Date signed

10/15/81

Date first disclosed to me

Signature of Witness

J. A. Cahill

Date signed

10/15/81

Date first disclosed to me

T. S. No. 8

Date Assigned 10/16/81

(1) Title of Invention Improved Process for the Production of Diteritary Butyl Peroxide

(2) - (3) - (4) - (5)

Based on past experience with acid resin catalysts for MTBE and SBA production, these catalysts tend to yield more moderate reaction rates at higher selectivity than concentrated sulfuric or other mineral acid. A large pore size resin would be preferable in this instance.

The reactor may be a multistage packed bed with interstage cooling or a multitube heat exchange design.

Since the purity requirements for a diesel fuel additive may not be stringent, Isobutane oxidate, consisting of near equimolar proportions of crude TBA and TBHP, may be used as reaction feed thus lowering raw materials cost.

The reaction is driven to completion and the resulting crude DTBP - water mixture may be washed with water or slightly alkaline solution to remove soluble formates, aldehydes and ketones originally present in the feed; trace acid from the resin catalyst is also washed or neutralized in this step. The DTBP is then phase separated and dried (if required). Because the reactor effluent does not contain sulfuric acid, the caustic quench and neutralization is eliminated.

Project No. _____		(6) Test data and original references may be found on the following notebook pages <u>187436</u> , <u>187437</u>	
Inventor (s) Signature <u>E. A. Hazbun</u> <i>E. A. Hazbun</i>		Date signed <u>10/13/81</u>	Date of Conception
Signature of Witness <u>V. M. Chong</u>		Date signed <u>10/15/81</u>	Date first disclosed to me
Signature of Witness <u>J. A. Cahill</u>		Date signed <u>10/15/81</u>	Date first disclosed to me

IMPORTANT

Technical Suggestion Form for attachment to the end of the notebook. (1) Title of the invention. (2) Your solution of the problem. (3) Your solution of the problem. (4) Give test results illustrating merits of your invention. (5) Describe principal advantages over prior practice which are realized by your invention. (6) List reference pages of laboratory notebook and the portions to whom the invention has been disclosed. (7) Names of inventors. (8) Cross out unused space. (9) Forward disclosure to Patent Department.

T. S. No. 81-7Date Assigned 10/10/81

(1) Title of Invention Improved Process for the Production of Ditertiary Butyl Peroxide

(2) - (3) - (4) - (5)

Such a process could yield DTBP of moderate purity at a transfer price in the 30 to 50¢/lb range depending on plant scale.

References

1. DTBP Production, R. L. Bobeck to B. Cramer, Aug. 1, 1979.
2. Initial Testing of DTBP as Octane Improver, J. M. DeJovine to J. A. Tarengelo, Jan. 14, 1981. Also U. S. Patent 2,378,341.

USE PEN OR TYPEWRITER - Suggested form for disclosure: (1) Title of Invention, (2) State the practices prior to your invention, the problems and connection with these prior practices, and how your invention solves these problems, (3) Your solution or invention - describe fully in prose, with chemical and structural formulae, when appropriate, (4) Give test results illustrating merits of your invention, (5) Describe principal advantages over prior practice realized by your invention, (6) List reference pages of laboratory notebook and the persons to whom the invention has been disclosed, (7) Names of inventors, witnesses are recorded on each sheet of the disclosure, (8) Cross out unused space, (9) Forward disclosure to Patent Department.

Project No. \_\_\_\_\_

(6) Test data and original references may be found on the following notebook pages 187437

Inventor (s) Signature

E. A. Hazbun

*E. A. Hazbun*

Date signed

10/13/81

Date of Conception

Date signed

Date of Conception

Signature of Witness

V. M. Chong

Date signed

10/15/81

Date first disclosed to me

Signature of Witness

J. A. Cahill

Date signed

10/15/81

Date first disclosed to me

IMPORTANT

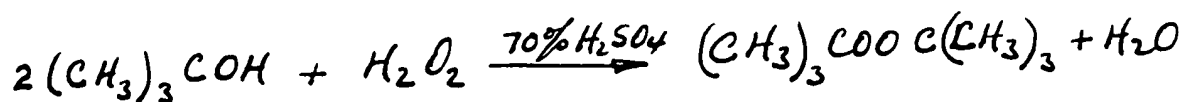
SUBJECT OF WORK

Improved process for the production of tertiary butyl peroxide.

T.S. 81-73 10/16/81 *void*

DATE

Ditertiary butyl peroxide (DTBP) is a specialty product used primarily as an organic peroxide catalyst or catalyst intermediate. It is commercially produced by the reaction of pure t-butanol (TBA) or isobutylene with hydrogen peroxide in the presence of concentrated sulfuric acid:



The process requires refrigeration to control the reaction isotherm ( $\Delta H = 98,000$  Btu/lb mole) at or below  $15^\circ\text{C}$  thus reducing side reactions and TBA dehydration. The product mixture is quenched with excess water to promote phase separation. The organic phase, consisting of mostly DTBP, is washed with dilute alkali solution and the aqueous acid phase is usually neutralized and discarded.

When excess hydrogen peroxide is used in the above reaction, a mixture tert-butyl<sup>hydro</sup>peroxide and DTBP ~~are~~ obtained.

SIGNATURE

L. F. Harbun

DATE

10/13/81

WORK CONTINUED ON PAGE

187435

WITNESSED AND UNDERSTOOD BY

*[Signature]*  
J. A. Cahill

DATE

10/15/81

10/15/81

Defect T.M. - Caalw - October 16, 1981

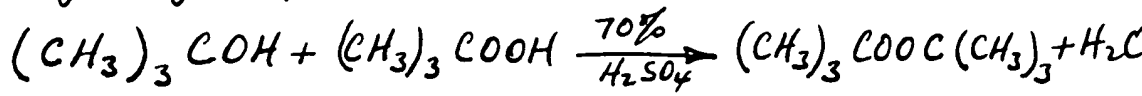
187434

OBJECT OF WORK Improved process for the production of  
ditertiary butyl peroxide

TS 81-73 10/16/81 cont'd.

DATE

An alternative process, that has been experimentally demonstrated and recently evaluated (1), substitutes tert-butyl hydroperoxide for the hydrogen peroxide:



This also requires refrigeration and acid neutralization. The estimated transfer price via this process at the 3 MM lbs/yr scale is \$1.16/lb <sup>for</sup> pure DTBP.

Recent preliminary testing of DTBP as a cetane improver for diesel fuels (2) have shown the following encouraging results:

V %	Cetane #
0	44.8
3	56.8
5	67.8

Such a performance may create a large volume application ~~for~~ DTBP if it can be manufactured at low cost.

SIGNATURE

E. J. Harburn

DATE 10/13/81

WORK CONTINUED ON PAGE 187434

WITNESSED AND UNDERSTOOD BY

N. M. Chong

DATE 10/15/81

J. A. Coyle

10/15/81

Yellowstone Co. Mc Carlin - October 16, 1981



OBJECT OF WORK

Improved process for the production of  
di-tertiary butyl peroxide.

T.S. 81-73 10/16/81 cont'd.

DATE

To allow continuous production at low cost, I propose that DTBP be produced by the reaction of TBHP and TBA over an ion exchange acid resin catalyst at a temperature sufficiently elevated to permit cooling with cooling tower water obviating the need for refrigeration.

Based on past experience with acid resin catalysts for MTBE and SBA production, these catalysts tend to yield more moderate reaction rates at higher selectivity than concentrated sulfuric or other mineral acid. A large pore size resin would be preferable in this instance.

The reactor may be a multistage # packed bed with interstage cooling or a multitube heat exchange design.

Since the purity requirements for a diesel fuel additive may not be stringent, Isobutane oxidate, consisting of <sup>near</sup> ~~about~~ equimolar

SIGNATURE

E. F. Hartman

DATE

10/14/81

WORK CONTINUED ON PAGE

187437

WITNESSED AND UNDERSTOOD BY

W. M. Chong

DATE

10/15/81

J. A. Cabell

10/15/81

Specult E. F. Hartman - October 16, 1981

OBJECT OF WORK Improved process for the production of  
di-tertiary butyl peroxide

T.S. 81-73

10/16/81 concluded

DATE

proportions of crude TBA and TBHP, may be used as reaction feed thus lowering raw materials cost.

The reaction is driven to completion and the resulting crude DTBP - water mixture may be washed <sup>with water or</sup> ~~in~~ slightly alkaline solution to remove soluble formates, aldehydes and ketones originally present in the feed; trace acid from the resin catalyst is also washed or neutralized in this step. The DTBP is then phase separated and dried (if required). ~~Since~~ Because the reactor effluent does not contain sulfuric acid, the <sup>caustic</sup> quench and neutralization is eliminated.

Such a process could yield DTBP of moderate purity at a transfer price ~~below~~ in the 30 to 50 \$/lb range depending on plant scale.

### References

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- 2- Initial testing of DTBP as Octane Improver, J.M. DeJovine to J.A. Tarengeloc Jan 14, 1981. Also US Patent 2,378,341

SIGNATURE

E. F. Hartman

DATE 10/14/81

WORK CONTINUED ON PAGE     

WITNESSED AND UNDERSTOOD BY

Arthur M. Chong  
J. A. Colwell

DATE

10/15/81

10/15/81

Oct 16, 1981 - Pelchert & McCallin